MAGNETIC PROPERTIES OF $NdCo_2Si_2$, $TbCo_2Si_2$ AND $DyCo_2Si_2$ IN HIGH MAGNETIC FIELDS*

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The magnetic properties of RCo_2Si_2 (R = Nd, Tb, Dy) compounds were studied by measuring the high-field magnetization in magnetic fields up to 140 kOe and over the temperature range 4.2–45 K. The magnetic field induces in $NdCo_2Si_2$ three metamagnetic phase transitions and two transitions in $TbCo_2Si_2$ and $DyCo_2Si_2$ compounds.

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1. Introduction

RCo_2Si_2 compounds have been investigated intensively over the last twenty years [1–3]. Problems of magnetic phase transitions in these materials are very interesting. For example, $PrCo_2Si_2$ exhibits three successive magnetic transitions below T_N of 30 K: commensurate long-pitch square-wave structures with propagation wave vectors along the c-axis, $k = 7/9$ for $T_2 < T < T_N$, $k = 13/14$ in the range of $T_1 < T < T_2$ and $k = 1$ for $T < T_1$, where $T_1 = 9$ K and $T_2 = 17$ K [3–6].

The magnetic properties of the other RCo_2Si_2 compounds have been studied in high-field magnetization measurements in magnetic fields up to 140 kOe at different temperatures. On the basis of these measurements the magnetic phase diagrams for some RCo_2Si_2 compounds were determined [7].

In this work new results of high-field magnetization measurements on $NdCo_2Si_2$, $TbCo_2Si_2$ and $DyCo_2Si_2$ compounds up to 140 kOe are reported.

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2. Experiments and results

Experiments were carried out on polycrystalline samples reported in the previous paper [3]. The samples were oriented in the magnetic field ($H = 100$ kOe). The magnetic measurements were carried out at low temperatures using a vibrating sample magnetometer in high magnetic fields up to 140 kOe in a "solenoid" installation. Results of magnetic measurements for $TbCo_2Si_2$, $DyCo_2Si_2$ and $NdCo_2Si_2$ compounds at high magnetic fields and different temperatures are shown in Figs. 1, 3 and 5. The results for the individual compounds are discussed below.

2.1. $NdCo_2Si_2$

An additional ac susceptibility measurement in a low magnetic field indicates the following magnetic phase transitions at temperatures of 15.5, 23.8 and 31.5 K. These values are in very good agreement with those observed for a single crystal [8].

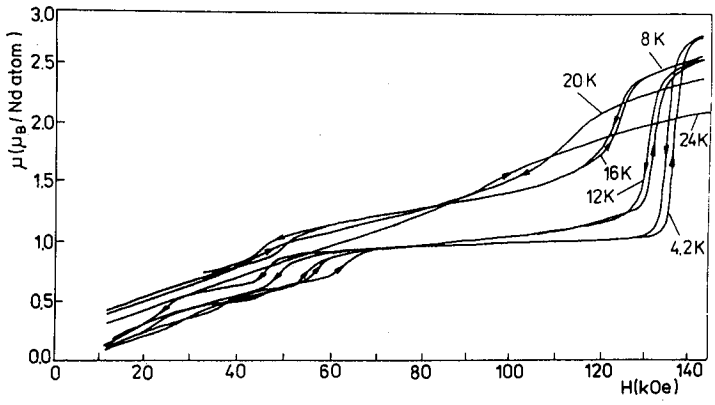

Fig. 1. High-field magnetization curves at different temperatures for $NdCo_2Si_2$.

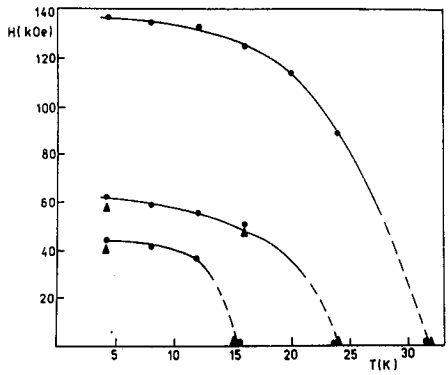

Fig. 2. Magnetic phase diagram for $NdCo_2Si_2$ (• — this work, full triangles — results taken from Shigeoka et al. [8]).

Figure 1 shows the magnetization curves at various temperatures. At $T = 4.2$ K, a step-like magnetization curve is observed with increasing field at $H_{c1} = 44$ kOe, $H_{c2} = 62$ kOe and $H_{c3} = 136$ kOe. The results reported previously [7, 8] indicate only the first two critical fields. The first and the second phase transitions have a large thermal anisotropy, while the anisotropy of the third is very small.

The magnetization at the maximum field $H = 140$ kOe reaches 2.8 $\mu_B/f.u.$ and it is smaller than the value for a free-ion Nd^{3+} moment ($gJ = 3.27\ \mu_B$).

An increase in temperature causes a change in the value of the critical field. Above $T = 30$ K the shape of the magnetization curve changes drastically. The magnetic phase diagram determined is presented in Fig. 2.

2.2. $TbCo_2Si_2$

Figure 3 shows the magnetization curves of $TbCo_2Si_2$ measured at different temperatures. The magnetization curves at $T = 4.2$ K reflect a two-step metamagnetic process with critical field $H_{c1} = 65$ kOe and $H_{c2} = 112$ kOe. In the temperature region 4.2–20 K the magnetization process is similar to that

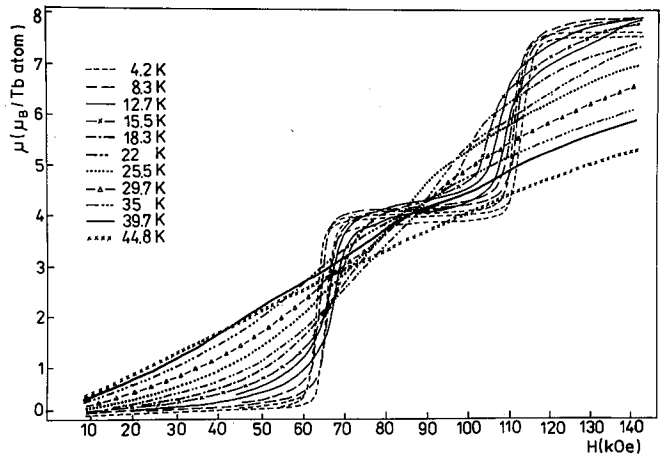

Fig. 3. High-field magnetization curves at different temperatures for $TbCo_2Si_2$.

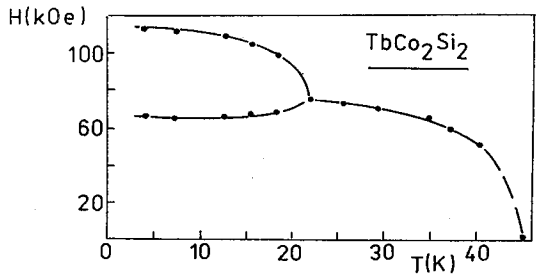

Fig. 4. Magnetic phase diagram for $TbCo_2Si_2$.

observed at $T = 4.2$ K. Above this temperature the magnetization process changes drastically, only one critical field is observed.

With magnetometric data the magnetic phase diagram was determined (see Fig. 4).

2.3. $DyCo_2Si_2$

Figure 5 shows the magnetization curves for $DyCo_2Si_2$. They have a similar character to those observed for $TbCo_2Si_2$ (cf. Fig. 3). The magnetization curves

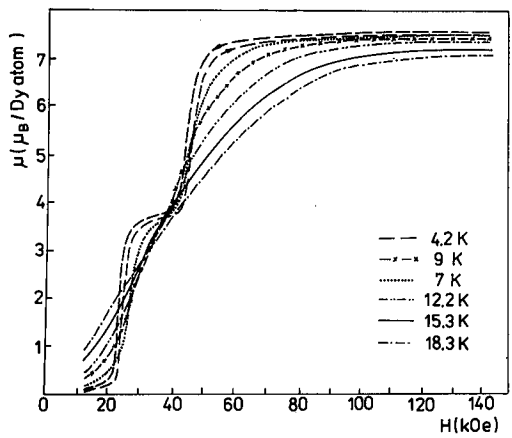

Fig. 5. High-field magnetization curves at different temperatures for $DyCo_2Si_2$.

at $T = 4.2$ K exhibit a two-step metamagnetic process with the critical fields $H_{c1} = 25$ kOe and $H_{c2} = 49$ kOe. The character of magnetization curves and values of critical fields are in agreement with the data for the single crystal $DyCo_2Si_2$ [9]. Below $T = 12$ K a two-step magnetization process is observed while above $T = 12$ K a one-step magnetization process is detected.

3. Discussion

The compounds investigated in this work crystallize in a body centred tetragonal structure of $ThCr_2Si_2$-type [10].

Neutron diffraction studies at zero magnetic field and $T = 4.2$ K indicate that in these compounds the magnetic moments localized only on R^{3+} ions form a simple antiferromagnetic structure AFI-type [11]. This type of magnetic structure could be displayed as a pilling up of the ferromagnetic sheets along c-axis with the sequence $+ - +-$ etc. and with the magnetic moment parallel to [001] direction.

In the case of $NdCo_2Si_2$ a collinear AFI-type structure is observed up to 15 K. Square wave with the propagation vectors $\boldsymbol{k} = (0, 0, 0.928)$ and $(0, 0, 0.785)$ appear for 15 K $< T <$ 24 K and 24 K $< T <$ 32 K $(= T_N)$, respectively [8]. The magnetic structure of $TbCo_2Si_2$ and $DyCo_2Si_2$ is of AFI-type and it is stable

in the temperature range 4.2 K$\div T_N$ [12–16]. Magnetic data for $NdCo_2Si_2$ presented in this work report for the first time the metamagnetic phase transition at $H_{c3} = 136$ kOe whereas other phase transitions were reported previously [7, 8]. These results indicate that magnetization curves for $NdCo_2Si_2$ have a three-step character. Two intermediate magnetic phases correspond to the incommensurate structures similar to those observed when the magnetization curves were measured as a function of temperature at zero magnetic field [8]. The $PrCo_2Si_2$ compound has also a multi-step magnetization curve [6]. The magnetization process for $NdCo_2Si_2$ and $PrCo_2Si_2$ compounds can be described on the basis of theoretical models developed by Date [17] and Iwata [18]. In these models, the Ising spins are immersed in the incommensurate sinusoidal exchange field $J(k) = J_0 \sin(kr + \delta)$, where r is taken along the c-axis and δ indicates the phase, and each spin points to the local field direction. These models give different incommensurate magnetic structures with different values of the wave vector in the function of magnetic field.

A different magnetic moment field dependence of the magnetization curves at low temperatures is observed for $TbCo_2Si_2$ and $DyCo_2Si_2$. The magnetization curves have a two-step character. Below H_{c1} the antiferromagnetic collinear structure is observed of AFI-type $(+-+-)$. In the intermediate region ($H_{c1} < H < H_{c2}$) the ferrimagnetic order (with the $++\,+-$ sequence) [9] or modulated one [19, 20] is observed, while for $H > H_{c1}$ the ferromagnetic ordering is stable.

The magnetic properties of these compounds may be described by the molecular field theory when the effective Hamiltonian includes exchange interactions, crystal field effects and interactions with external magnetic field. The theoretical investigation by Katsura and Narita [21] shows that we should take J (exchange integral) at least up to $n = 3$ to ensure the appearance of the structure with $++\,+-$ sequence in the intermediate region.

With increasing temperature a change in character of the phase transition is observed. The complicated magnetic behaviour observed in these compounds may be caused by a competition between exchange interactions and strong crystalline electric field effects.

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